

Coulomb-Blockade Devices as a Microscope of Low-Temperature Dynamics in Disordered Materials

William H. Huber and Neil M. Zimmerman¹

neil.zimmerman@eeel.nist.gov; <http://www.eeel.nist.gov/811/femg/set.html>

National Institute of Standards and Technology²
Gaithersburg, MD 20899

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Abstract

We report systematic measurements of the charge offset in submicron-sized single-electron tunneling (SET) transistors. In some devices, we observe a monotonic transient relaxation in the rate of drift of the charge offset over days or weeks. We present an extension of the amorphous material two-level systems model (previously measured only in macroscopic samples) which naturally explains the drift as arising from atomic structural relaxations. We point out that this result opens a new “microscope” into atomic-scale dynamics, and has implications for the error rates in SET pumps.

¹to whom correspondence should be sent

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The Coulomb blockade^[1] is a phenomenon based on the non-zero energy necessary to charge a capacitor with one more electron; the minimum energy is $\sim e^2/C$. One of the consequences of this is that it is possible to make a single-electron tunneling (SET) transistor (SETT) which is sensitive to changes of charge of about $10^{-3} e$. This exquisite charge sensitivity (several orders of magnitude better than any other technology) forms the basis of an ultra-sensitive charge electrometer; unfortunately, it also means that the transistor is sensitive to trapped charges in the areas of insulator which typically reside in a thin film-based device.

This “charge offset”, denoted by the displacement charge Q_0 on the SETT island, is a problem in several ways. For one, the time-dependent fluctuation of Q_0 limits the resolution of the charge electrometer. Quite a few studies^{[2][3][4][5][6]} have been performed on the short-term (typically 1/f) noise, usually in the range of 0.1 to 1000 Hz. The total change in Q_0 due to noise in this range is typically of order $0.01 e$ or less. This small change implies that measurements of the 1/f noise are important for the sensitivity of the SETT as a charge electrometer, but they don’t impact on the ability to integrate SET devices.

In contrast, it has been observed that, for much longer times (or due to thermal cycling or other external perturbations) Q_0 can change by a large fraction of e , and can thus significantly change the behavior of the device (in the parlance of digital circuits, the Q_0 change can turn a device from “on” to “off”, or vice versa). The charge offset is thus an important and oft-discussed^{[7][8]} problem in the SET field, but no systematic experimental

studies have been performed. One of the results of our work is that the short-term $1/f$ noise may not be correlated with the long-term drift that can change Q_0 by large fractions of e ; thus, the short-term noise does not appear to be a useful predictor for the integrability of recently-fabricated devices.

Another experimental observation which has been anecdotally reported fairly often is that the noise (short- or long-term) in SETT's decreases in a monotonic way for a few days or weeks after the device is cooled; however, to our knowledge no systematic studies have been pursued. In this paper, we report systematic studies of the long-term (periods from a few minutes to many weeks) drift in Q_0 . In particular, we focus on one striking result: in some devices, the long-term drift has a “transient relaxation”, i.e., a systematic and monotonic decrease in the rate of drifting, over the first few days or weeks after fabrication or cooldown. This relaxation bears a striking similarity to an old result from a very different field: the non-equilibrium heat evolution observed in glasses (e.g., silica) when quenched to temperatures below 1 K.

The study of the low-temperature properties of amorphous materials has a long history^[9]; the anomalous (different from crystals) thermal and acoustic properties at temperatures below a few K have been attributed to a model of “two-level systems” (TLS), in which the dynamics are driven by structural reconfigurations of a set of microscopic entities which have two possible states. In general the identity of the TLS are unknown, but typically believed to be complicated reconfigurations of a few (of order ten to one hundred) atoms.

One of the observations in highly disordered materials such as silica^[10] is that, below about 0.3 K, there is a long-time tail thermal relaxation; specifi-

cally, on quenching from higher temperatures, the glass releases heat with a thermal power that decreases as $1/t$, where t is the time since quench. This heat evolution can be explained naturally in the TLS model, as due to TLS which are frozen into non-equilibrium energy states by the quench, and are slowly relaxing to the ground state while releasing energy.

If there is disordered material in the insulating regions surrounding the thin film SETT, the structural relaxation – if it is accompanied by a change in the charge distribution of the TLS (as might occur as the atoms move around) – can give rise to a change in Q_0 , with a rate of drift $\delta Q_0(t) \propto 1/t$. We will show that a simple extension of the TLS theory is in quite good agreement with all of our data, including temperature and time dependence of both the long-term drift and the short-term noise, as well as the full frequency dependence over the entire range of times. We also note that there is one disagreement: the transient relaxation in $Q_0(t)$ does not recur with thermal cycling; we discuss later this one disagreement and its effect on our conclusions.

We fabricated standard three-terminal SETT's with Al/AlO_x/Al tunnel junctions (Fig. 1C inset) using a process^[11] similar to the standard double angle-deposition technique.^[12] On some of these devices, we put an extra layer about 300 nm thick (the “blanket”) of amorphous AlO_x (Fig. 1A inset). We note that the tunnel junctions are made by room-temperature oxidation of Al, whereas the blanket is made from an electron-beam evaporation of amorphous alumina in the presence of 10^{-5} Torr of oxygen. Typical base pressure was 1×10^{-7} Torr, deposition 3 to 10×10^{-7} Torr. For the devices represented here, the devices were cold (4 K) within 1 1/2 days after

deposition.

Fig. 1A shows the time dependence of the charge offset $Q_0(t)$ for the blanketed device; we measured this by repeatedly sweeping the gate voltage (on the lead at the top of the inset), and then fitting the oscillations in source-drain current I_{S-D} (horizontal flow in the inset) to determine the phase shift. Note that $Q_0 = 0$ corresponds to minimum current (maximum blockade), and that, since Q_0 is only definable modulo $1e$, the upper and lower edges of the graph ($0e$ and $1e$) wrap around.

This figure shows the *main experimental result of this work: a strong, systematic decrease in the rate of drift of $Q_0(t)$* . Initially, the magnitude of the time rate of change of the charge offset (i.e. $|dQ_0(t)/dt|$) is so large that subsequent measurements of Q_0 (spaced five minutes apart) appear random. After several days, $|dQ_0(t)/dt|$ decreases enough to clearly detect clustering of Q_0 . We find that $|dQ_0(t)/dt|$ continuously decreases over time; thus after three weeks Q_0 remains relatively stable for several days. It is also noticeable in this device that $dQ_0(t)/dt$ rarely changes sign; this monotonic trend in $Q_0(t)$ is sometimes, but not always, present in the measured transient relaxation of devices.

Fig. 1B shows the amplitude of the power spectral density $S_Q(f)$ of Q_0 at 10 Hz for the same device, measured over the same three weeks; gaps in the data represent periods during which extrinsic experimental difficulties prevented the noise measurement. $S_Q(f)$ was measured by a spectral analysis of the time dependence of the I_{S-D} . In general, $S_Q(f) \propto 1/f$, with an absolute amplitude ($10^{-6}e^2/\text{Hz}$ at 10 Hz) similar to other measurements. *We note that, in contrast to the long-term drift $Q_0(t)$, the amplitude $S_Q(f)$*

does not decrease over the same time period, (within uncertainty of about 20%).

Fig. 1C shows a similar micrograph and $Q_0(t)$ for an *unblanketed device*. We note that there is no indication of a transient relaxation noticeable in the time record.

Fig. 2 shows a slice of Figures 1A and 1B, during which the temperature T was elevated, including $T = 1.5$ K between 8.8 and 9.1 days. We see quite clearly that the short-term $1/f$ noise (lower panel) responds to these changes; $S_Q(f)$ increases with T , as expected.^[3] In contrast, the rate of the long-term $Q_0(t)$ drift (upper panel) did not respond at all to the change in T .

We can quantify the transient relaxation through the “rate of drifting,” by repeatedly measuring how long it takes for $Q_0(t)$ to change by more than a given amount, bin those duration times, and measure the characteristic time for that Markov process. Such an analysis is represented by Fig. 3, which shows the rate γ (the inverse of the characteristic time) as a function of the running time, for both devices in Fig. 1. We note that this procedure can reveal a fairly weak change in γ ; both devices show some transient relaxation, with the blanketed device being much stronger. The inset shows a plot of γ versus $1/\text{time}$ on a log-log plot; *it is quite clear that the rate of long-term drift in $Q_0(t)$ is inversely proportional to time since the deposition*. The observations embodied in Figures 1A and 3 are very reminiscent of the non-equilibrium heat evolution in amorphous materials.

Given this empirical similarity, can we use the TLS model to predict the absolute magnitude, and temperature and time dependences of γ ? We use the “standard” TLS model; following Black^[13], the time-dependent distribution

of TLS *which have already relaxed and emitted energy E* is

$$n(E, t) = (P(E)/2) \ln(4t/T_{1,\min}(E)),$$

where E is the total energy splitting, t is the time since cooldown, $P(E)$ is the “universal” TLS density per energy and volume (Black uses the average \bar{P}), and $T_{1,\min}$ is the relaxation time for $\Delta_0 = E$, where Δ_0 and E are the tunneling and total energies.

We can extend this model as follows: first, the rate of release of energy packets between E_{\min} and E_{\max} (V is sample volume) is

$$\begin{aligned} \dot{N} &= V d/dt \int_{E_{\min}}^{E_{\max}} dE n(E, t) = V \int_{E_{\min}}^{E_{\max}} dE dn(E, t)/dt. \\ \dot{N} &= V/(2t) \int_{E_{\min}}^{E_{\max}} dE P(E). \end{aligned} \tag{1}$$

We note that this rate does not depend on temperature, and is indeed inversely proportional to time (both in agreement with the data, as in Figures 2 and 3).

Finally, to get a numerical estimate for \dot{N} , we must define E_{\min} , E_{\max} , and $P(E)$. To do this, we must first consider the one significant difference between our results and the typical heat evolution measurements: as mentioned in the introduction, the transient in $Q_0(t)$ does not recur on thermal cycling, but instead occurs only once, apparently after device fabrication. We believe that this is because, unlike silica, thin film-based AlO_x is not intrinsically amorphous; rather, it has a great deal of structural disorder and in-plane stress by virtue of its fabrication. We speculate on two specific mechanisms that could lead to the non-recurrence: It seems likely that the

as-deposited film stress will relax as a function of time since deposition, but will not recur on cycling (as seen in $Q_0(t)$). Also, it is known that both 1/f noise in thin metal films ^{[14][15]} (not SET devices) and low-temperature TLS dynamics^[16] can arise from specific defect mechanisms that do not have broad energy distributions ranging down to $E = 0$. If this is the case for the defects causing the transient, then thermal cycling up to room temperature may not be enough to re-populate those defect TLS.

Given this surmise, we can now use Equation 1 to estimate the rate of drift by assuming that, in these alumina films, there is the same total density of TLS (for all E , per unit volume) as in truly amorphous materials. We thus replace $P(E)$ by the average \bar{P} deduced from heat evolution measurements^[10], E_{\min} by 0, and E_{\max} by 20 K^[17]. Finally, we assume that every relaxation (decrease in $n(E, t)$) results in an electrostatic atomic reconfiguration which changes Q_0 (and thus contributes to γ), and that the site of the TLS relaxations which cause changes in Q_0 is the AlO_x in the SETT tunnel junctions or on the surface of the Al island (a volume V made of a “skin” of area $0.5 \mu\text{m}^2$ and thickness of 100 nm). We obtain

$$\begin{aligned}\gamma &\approx 1300/t \\ &\approx 2/\text{hour (after three weeks)}\end{aligned}\tag{2}$$

Thus, three weeks after the cooldown, we expect a significant change in Q_0 about once every hour. This rate is about a factor of ten higher than in Fig. 1A; given the crude approximations, and especially the large extrapolation from the calorimetric measurements of heat evolution on cm^3 of material to submicron thin film samples, this numerical agreement with the experiment

seems quite compelling. Also, we can see that, in agreement with Figures 1A, 2, and 3, the transient relaxation in the rate of the long-term drift should be inversely proportional to time, but should not depend on temperature.

Of course, the short-term 1/f noise dependences in Figures 1B (time dependence) and 2 (temperature dependence) do not agree with these qualitative predictions. We can naturally explain this in the context of the same model, by adding a second, incoherent noise source: in addition to the long-term drift from the non-equilibrium relaxation, we add the standard assumption of thermal equilibrium 1/f noise. In particular, we can predict a power spectral density of charge fluctuations spanning the whole range of times between short- and long-term: we add incoherently a 1/f noise with amplitude $10^{-6}e^2/\text{Hz}$ at 10 Hz to a Lorentzian^{[18][19]} $S_Q(\omega) = (\delta Q_0^2/2\pi\gamma)[1/(1 + (\omega/2\gamma)^2)]$. Here, γ is from Equation 2, and δQ_0 is the amplitude of the fluctuation in Q_0 for each structural relaxation, which we approximate as $0.1e$.

Fig. 4 shows these theoretical predictions as the dotted (non-equilibrium noise) and dashed (equilibrium 1/f noise) lines. The vertical dashed line indicates where these two different types of noise overlap. This prediction yields exactly the qualitative dependences seen in the data: at low frequencies (corresponding to the long-term drift), the noise is dominated by the non-equilibrium noise, which is time dependent but temperature independent. At high frequencies (the short-term regime), the noise is equilibrium thermally-excited 1/f noise, which is temperature dependent but stochastic (not time dependent).

Unfortunately, experimental limitations prevented simply measuring $S_Q(f)$

over this entire frequency range. However, we can obtain selected ranges: at frequencies near 10 Hz, we have measured $S_Q(f)$ (as shown by squares in Fig. 4), from the power spectral density of $I_{\text{S-D}}$. At low frequencies, we have numerically obtained the spectrum from $Q_0(t)$ in Fig. 1A, for a time near the end of the three weeks (the upper limit of 1 mHz is set by the time between successive measurements of about five minutes). We can see that both ranges of measurements above 0.1 mHz agree quite well with the predictions of the incoherently summed $1/f$ and relaxation (no fit parameter) noises. The exception is the lack of a strong rolloff at very low frequencies (less than 0.1 mHz), indicating the actual rate γ is smaller than predicted than predicted by Equation 2, as noted above.

While the quantitative agreement is appealing, we believe that the compelling part of this agreement is as outlined above: the transient relaxation in the long-term drift and the short-term $1/f$ noise have different qualitative behaviors because they have different origins.

We note that most of the main conclusions in this paper, namely 1) the experimental dependences of both long-term drift and short-term noise on time and temperature, 2) the corresponding qualitatively different origins of these two, and 3) the implications for error rates in pumps (see below), are independent of the numerical estimate in Equation 2, and thus of the specific TLS model used.

Finally, we comment on one important implication of the agreement between experiment and theory in this work. An important application for SET devices is that of using SET pumps^[1] for a standard of charge^[20] or current. A perplexing problem is that the error rate (more or less electrons

pumped than desired) is many orders of magnitude higher than predicted by theory.^[21] These errors, unlike the charge changes considered herein for SETT's, require an energy input equivalent to about 5 K.^[22] One possible explanation^[23] is that non-equilibrium relaxations, which could both provide energy and also induce voltage fluctuations via the charge coupling, could be the cause. This explanation is supported by a numerical correlation between the observed error rates and an extrapolation of the measured $1/f$ noise to 100 GHz.

We believe that, for two reasons, this explanation may be suspect. First, on energy grounds, the model considered herein clearly demonstrates that, within a few days after cooldown, for glassy volumes of order μm^3 , there are not enough energy relaxations to drive the SET pump errors. Secondly, the model and data in Fig. 4 suggest strongly that, given the pump errors require energy input and therefore correspond to the non-equilibrium dotted curve which falls as $1/f^2$ above 1 mHz, an extrapolation based on a $1/f$ spectrum markedly overestimates the spectral density at 100 GHz.

To summarize, we have performed systematic measurements of the long-term drift of the charge offset in SET transistors. We have observed that there is a transient relaxation in the drift in some cases. The qualitative temperature and time dependences are different from those of the short-term ($1/f$ -like) noise. We have demonstrated that an extension of the TLS model for glasses and amorphous materials can explain the quantitative and qualitative behaviors of the drift as driven by non-equilibrium relaxation of the disordered atoms. By adding in $1/f$ noise, we can also explain comprehensively the behaviors of both the short-term noise and long-term drift; this

suggests that using only the short-term noise to evaluate the integrability of SET devices may be inadequate. This work also has implications for several applications of SET devices, including error rates in SET pumps.

Finally, we note that this measurement opens up a new “microscope”, which allows examination of low-temperature atomic dynamics on thin film-based sub- μm samples.

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List of Figures

1. 1A: Inset: Micrograph of “blanketed” SETT. White areas are Al on a Si chip; central square is overlayer of alumina. Data shows the long-term drift of the charge offset $Q_0(t)$, measured over about three weeks; horizontal axis is time in days since deposition. This graph shows clearly the “transient relaxation”, i.e., the systematic decrease in the rate of drift of $Q_0(t)$. “A” and “B” denote a range of time during which the temperature was elevated, as high as 1.5 K. At longer times, the drifting in $Q_0(t)$ seems to become stationary, appearing similar to that in Fig. 1C

 1B: Time dependence of $1/f$ noise amplitude $S_Q(f)^{1/2}$ at 10 Hz, as a function of time, for blanketed device. Note that, unlike $Q_0(t)$, there is no transient in the short-term noise.

 1C: Micrograph and $Q_0(t)$ for SETT without any extra alumina. $Q_0(t)$ clearly shows a markedly different behavior, with very little transient relaxation.
2. An expansion of Figures 1A and 1B, showing the dependence, in the blanketed device, of charge offset drift (upper) and short-term noise (lower) during times when the temperature T rose as high as 1.5 K. The short-term noise has a strong dependence on T, but the long-term drift does not.
3. Main: Log-log plot of the rate versus $1/\text{time}$ for the blanketed device, showing an inverse linear time dependence. Inset: Dependence of the

rate of drift γ on time since deposition, for both devices shown in Figures 1A and 1C. Note that the blanketed device has a much stronger transient relaxation.

4. Power Spectral density of charge fluctuations versus frequency $S_Q(f)$; note that the noise Figures 1B and 2 plotted the square root of this quantity. Dotted line: prediction of extension of TLS model for non-equilibrium glass relaxation noise. Dashed line: prediction for standard $1/f$ noise. Vertical line: Below about 0.1 Hz, the non-equilibrium noise dominates; above 0.1 Hz, the $1/f$ noise does. Squares: (low frequency) numerical calculation of $S_Q(f)$ from $Q_0(t)$ in Fig. 1A (days 15 to 17); (high frequency) from real-time measurement of $I_{S-D}(t)$. The lack of a low-frequency rolloff in the data below 0.1 mHz is due to a difference between the predicted and measured rates.

Fig 1A
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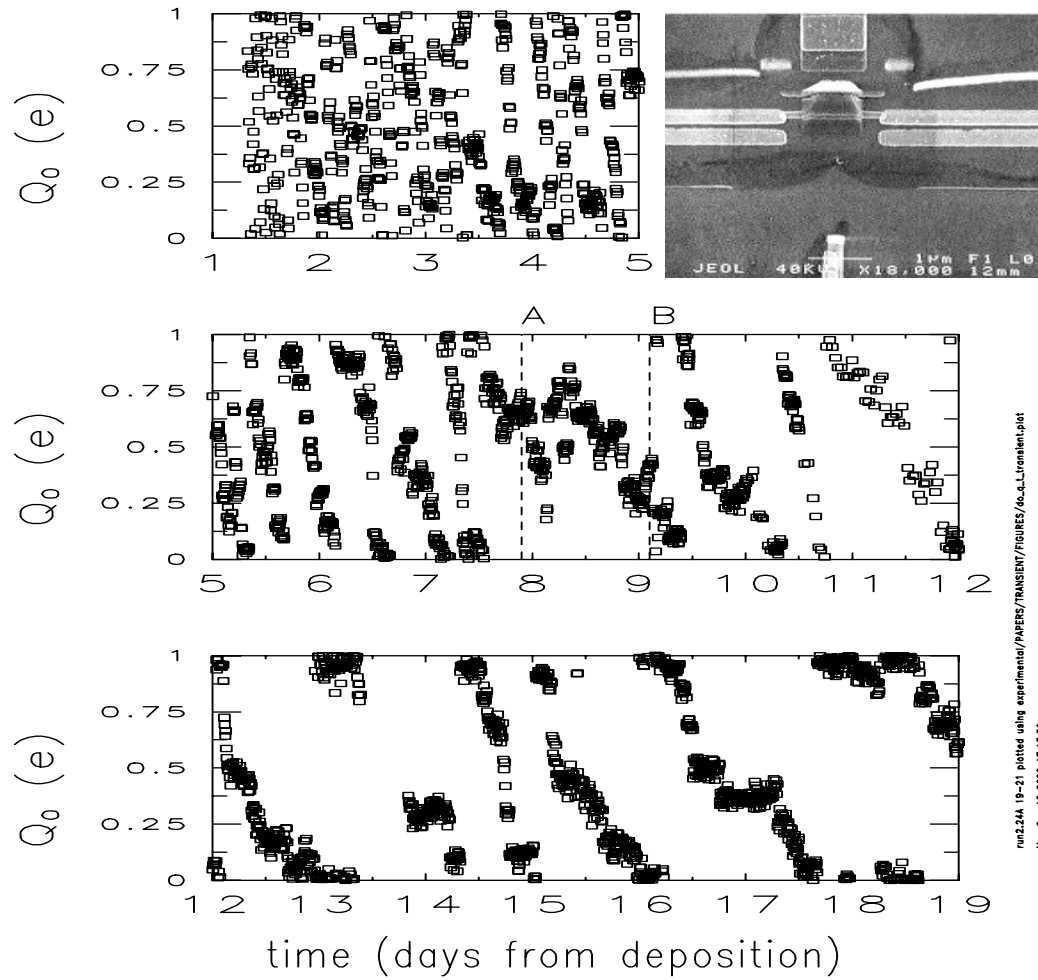
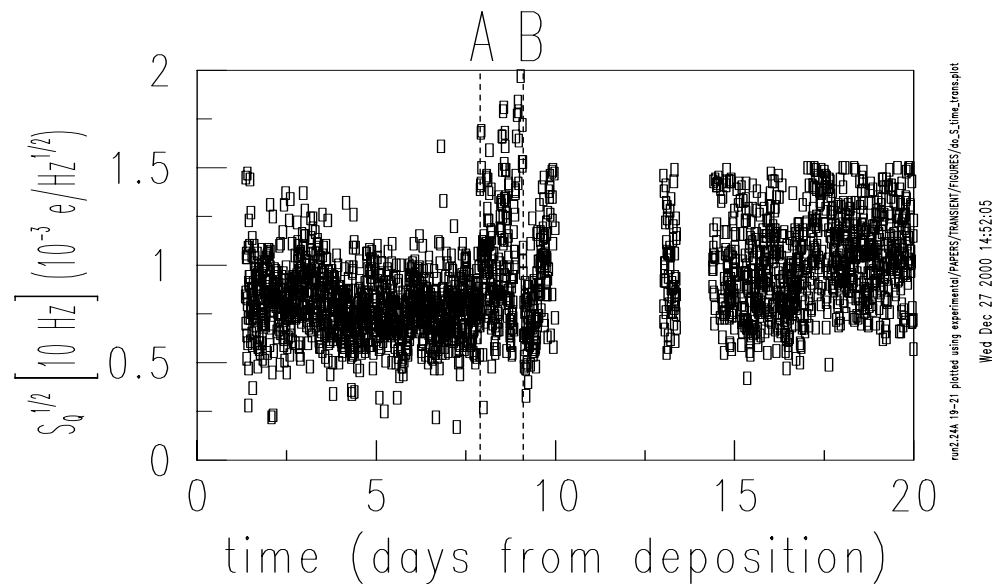


Fig 1B
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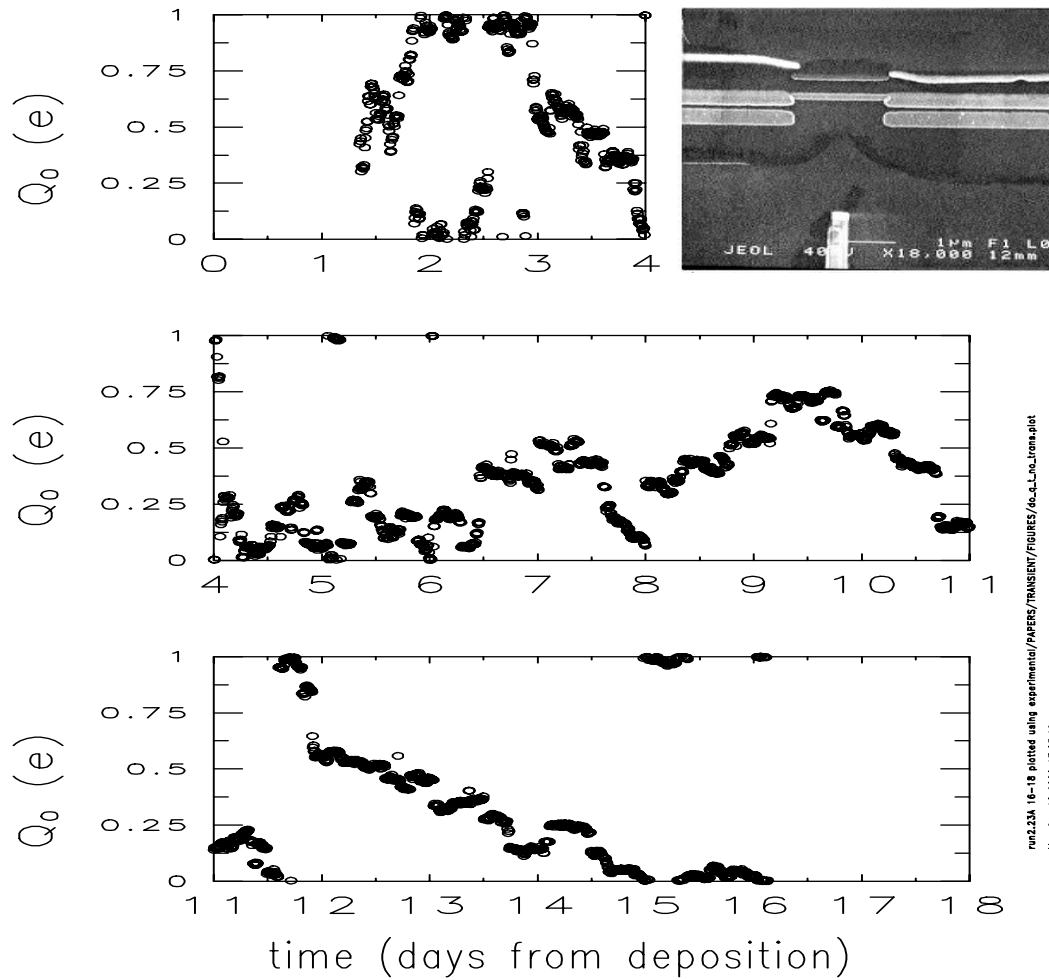


Fig 1C
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Fig 2
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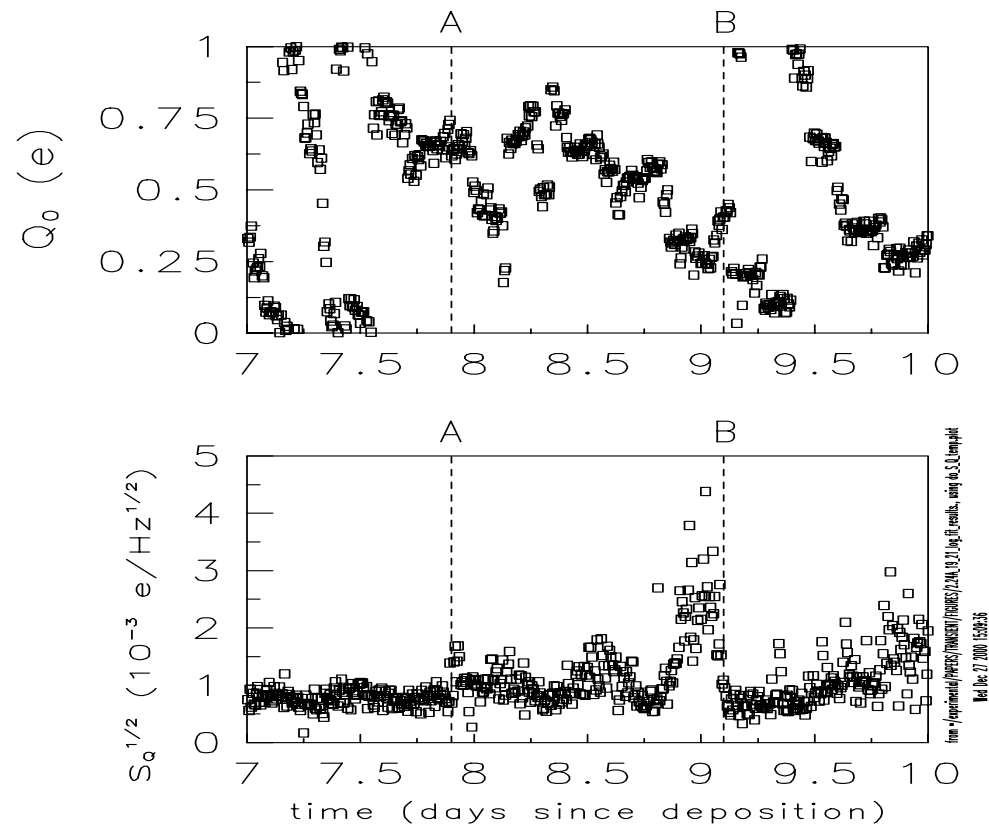
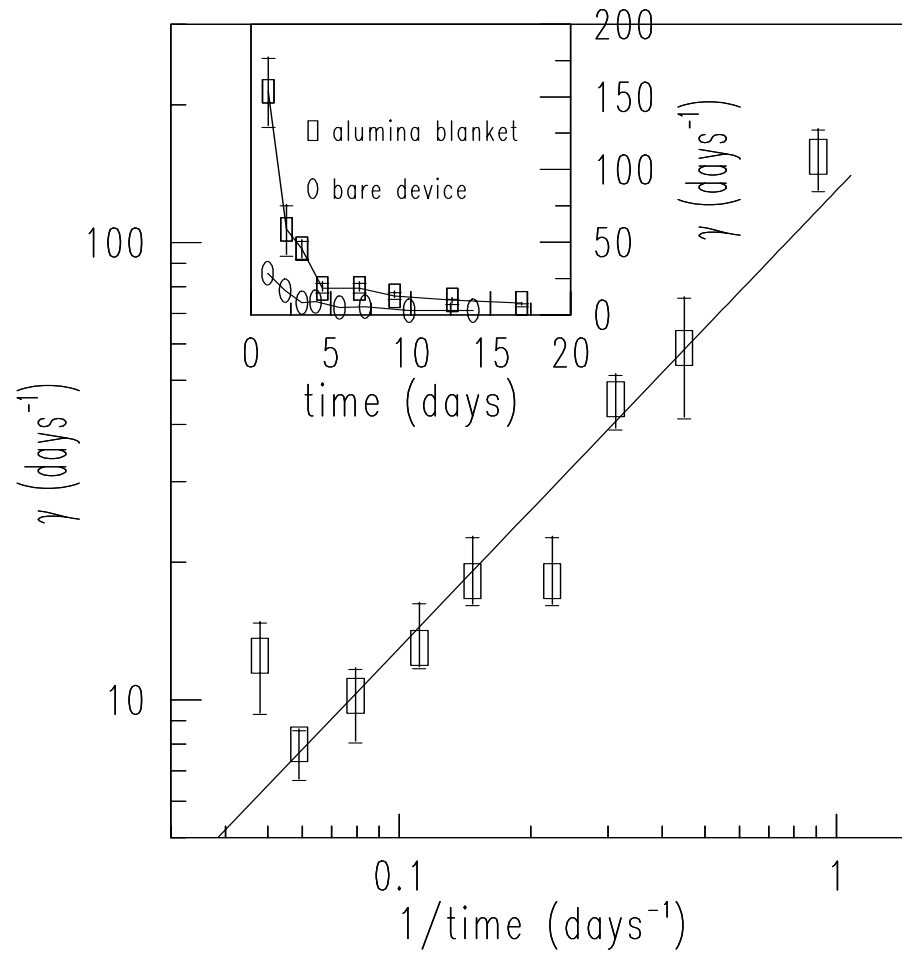


Fig 3
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run2.24A 19-21 plotted using experimental/PAPERS/TRANSIENT/FIGURES/do_gamma.t.plot
Tue Oct 31 2000 16:10:31

Fig 4
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